Fluctuation Kinetics of Diffusion-Controlled Processes: Strong Effects Due to Correlations and Fluctuations

S. F. Burlatsky[1] **and G. S. Oshanin**[1]

We summarize some recent results related to fluctuation-induced kinetics of diffusion-controlled processes. We show that kinetic behavior can be drastically changed due to fluctuation effects, spatial correlations between particles, and anomalous transport properties. In addition, we show that correlation-induced kinetics in some systems can govern the temporal evolution over the entire time domain.

1. INTRODUCTION

The study of many-particle aspects of diffusion-controlled processes (DCP) has recently attracted considerable attention.[1-6] This is due, on one hand, to a close relation to fundamental problems of statistical physics in which fluctuation-induced behavior is essential, and on the other hand, to a wide variety of applications ranging over an incredibly broad spectrum—the recombination of ions and defects in solids, energy transfer in liquid and solid solutions, chemical reactions, electron dynamics in disordered systems of repulsive scatterers, biological processes with migration, annihilation, and multiplication, and transport processes in disordered matter.[1-6]

This review is organized as follows: in Section 2 we briefly review some results based on the Smoluchowski mean-field approach. In Section 3 we present exact results concerning the influence of spatial fluctuations on the long-time reaction kinetics in some simple systems. In Section 4 we discuss the reaction kinetics in systems where the spatial fluctuations are decisive

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[1] Institute of Chemical Physics, USSR Academy of Sciences, Moscow V-334, USSR.

from the earliest times and govern the conversion of the bulk of reactive species. The latter are, e.g., systems with a high density of reactants or with correlations in the distribution of reactants, e.g., polymers, systems with an external particle random input or reproduction of the active species, solids with topological defects, and random systems with considerable disorder or distant reactions, in which random motion is not diffusive. Finally, in Section 5 we present some new results concerning non-Fickian fluctuation-induced transport behavior in disordered membranes.

2. DCP theory describes the kinetic behavior of many-particle processes, such as, e.g., the reactions

$$A + B \rightarrow C \tag{1a}$$

$$A + A \rightarrow C \tag{1b}$$

$$A + T \rightarrow T \tag{1c}$$

involving diffusive particles A, B, C [reactions (1a), (1b)] and diffusive or immobile traps T [reactions (1c)]. Two particles react when their distance is equal to R, which is termed the reaction radius. This distance is taken equal to the sum of the particle radii $R = R_A + R_B$ (or R_T) for contact reactions. The direct reaction, the formation of the reaction product C or capture of A in Eq. (1c), occurs with a finite probability which determines the direct rate constant K. The backward reaction, unimolecular breakup of C (release from the trap), is defined by the intrinsic constant K_-.

The field of DCP was initiated in the work of Smoluchowski[7] concerning the kinetics of irreversible coagulation of colloid particles. His approach, based on the single-particle diffusion equation with an absorbing boundary, leads to the following mass equation for the mean densities of particles:

$$\dot{C}_A(t) = \dot{C}_B(t) = -K_{\text{eff}} C_A(t) \, C_B(t) \tag{2a}$$

where the overdot denotes time derivative and the "effective" rate coefficient in three-dimensional systems is given by (for $4\pi DR \ll K$)

$$K_{\text{eff}} = 4\pi DR[1 + R/(\pi \, Dt)^{1/2}], \qquad D = D_A + D_B \tag{2b}$$

The Smoluchowski approach is the basis of traditional mean-field methods in DCP investigations.[1–3] In particular, K_{eff} has been evaluated for the case $4\pi DR \gg K$ (see ref. 8). A representation analogous to Eq. (2b) was derived for the effective constant of the backward reaction[9,10] in reversible reactions. Much emphasis has been put on studying the reactions (1), kinetics in low-dimensional systems $(d \leqslant 2)$, and in systems with nonlocal

interactions between species. It was shown that in low-dimensional systems the effective rate constant becomes an effective rate coefficient, i.e., K_{eff} becomes a function of time due to the peculiar properties of low-dimensional random walks. The potential interactions between reactive species does not affect the long-time dependence of $C(t)$, but lead to a renormalization of the effective rate constants within the framework of mean-field descriptions[10-12] and to a drastic change in the kinetic laws at short times.[13]

   3. The Smoluchowski approach has a major limitation since it ignores the many-particle nature of DCP problems. It is easily shown that it gives a lower bound for the mean concentrations.[14] The course of any reaction leads to the appearance of spatial correlations in the distribution of reactants. Within the Smoluchowski approach only the short-wave two-particle correlations are taken into account, but long-range many-particle correlations are neglected, and the independence of reaction probabilities for each reacting pair is assumed.

   It is worthwhile to mention that for a wide variety of many-particle problems in statistical physics the mean-field approximations are quite acceptable. In contrast, problems in which many-particle behavior is significant are, as a rule, unsolvable. However, within the last years there has been considerable progress in developing a many-particle description of DCP kinetics. It has recently been shown that a great majority of DCP are correlation-controlled in the large-t limit and, at the same time, a large body of exact results has been obtained.[14-17] In particular, it has been shown that the kinetics of the trapping reaction in Eq. (1c) at $t \to \infty$ depends drastically on the spatial distribution of traps T. For a random placement of traps the Smoluchowski approach, which predicts an exponential decay of A, is invalid and the trapping kinetics exhibits the unusual behavior[14-21]

$$\ln C_A(t) \propto -t^{d/d+2} \tag{3}$$

This stretched-exponential decay stems from the presence of large, but very rare, trap-free cavities. In such a cavity the particle lifetime is anomalously long and the contribution of these long lifetimes leads to the decay law in Eq. (3). It is worthwhile to mention that Eq. (3) is similar (up to the inverse Laplace transform) to the well-known Lifschitz[22] result concerning the low-energy spectrum of an electron in an array of randomly distributed immobile scatterers. In the seminal paper on the subject[18] this law was evaluated as a lower bound on the survival probability, then an asymptotically exact result was derived,[19] and finally, corrections to the terms in Eq. (3) were calculated.[14]

The second important instance in which spatial correlations are important is the strictly bimolecular (SB) irreversible reaction in Eq. (1a) with $C_A(0) = C_B(0)$. It was shown in refs. 20 and 23–25 that the mean concentrations of A and B are characterized by an anomalous long-time decay

$$C_A(t) = C_B(t) \propto t^{-d/4} \tag{4}$$

which displays a slower kinetic behavior for $d < 4$ as compared to the mean-field predictions. The point is that the difference $z(r, t) = C_A(t) - C_B(t)$ induced by initial small fluctuations in the concentrations of A and B is not affected by the recombination reaction, because A and B particles disappear only in pairs. The diffusive decay of $z(r, t)$ is a slower process than the mean-field recombination decay of Eq. (2). For large t the reaction bath becomes separated into large domains (of characteristic size $R \propto t^{1/2}$) containing particles of only one type, and the kinetic behavior is controlled by the diffusive "transport" of A and B from regions enriched by A species and ones enriched by B into one another, which leads to the decay kinetics of Eq. (4). With the help of such qualitative arguments Ovchinnikov and Zeldovich[20] presented the following simple estimate:

$$C_{A,B}(t) = \tfrac{1}{2} \langle |z(r, t)| \rangle \propto t^{-d/4} \tag{5}$$

The exact equation for the evolution of the mean density contains the pair correlation functions[23]

$$\dot{C}_{A,B}(t) = -k[C_{A,B}^2(t) + \langle \delta C_A(r, t)\, \delta C_B(r, t) \rangle] \tag{6}$$

The special type of decoupling based on fourth-order correlations leads to the result[23]

$$C_{A,B}(t) = [-\langle \delta C_A(r, t)\, \delta C_B(r, t) \rangle]^{1/2} \propto t^{-d/4} \tag{7}$$

It is known that the estimate in Eq. (5) is a lower bound on the mean concentration,[14] while the estimate in Eq. (7) gives an upper bound. These inequalities [and the monotonic behavior of $C(t)$] strongly suggest that the fluctuation-induced asymptotic laws of Eqs. (5) and (7) are asymptotically exact. A similar proof was published by Bramson and Lebowitz in a set of fundamental papers.[15,16]

We mention that if the initial A and B densities are not equal to each other, correlation effects are suppressed at large times and the mean-field exponential decay is valid.[14–16,5] The mean-field description is also valid for the SB irreversible reaction of charged species.[26] Spatial regions created by density fluctuations induce local electric fields which smooth the initial thermal inhomogeneities faster than diffusion.

For the reversible reactions (1) there always exists a linear combination of local concentrations not affected by reaction due to the mass conservation law. In consequence, a wide variety of reversible bimolecular reactions are defined by a power-law approach to equilibrium at $t \to \infty$ [27–31,5] in place of the exponential mean-field decay law. For example, such kinetic behavior is exhibited by the reversible binary reaction[27] in Eq. (1b) or more general coagulation/fragmentation reactions[30] and the relaxation of the short order parameter in solutions.[5] A power-law decay defines the long-time kinetics of the strictly bimolecular reversible reaction in Eq. (1a), of uncharged species,[27,28] nonstoichiometric reactions[31] in (1a), and reactions (1a) that include charged species.[29,30] Interestingly, in the latter case the long-time decay is induced by a special type of density fluctuation, in which fluctuations have the same sign, i.e., the occurrence of regions in which deviations from the mean density have the same value and sign both for A and B. These electrically neutral fluctuations are smoothed only by diffusion, which causes a power-law approach to the equilibrium.[29,30] If the initial and steady states are described by a flat fluctuation spectrum, then the mean concentration relaxation is defined by

$$C_A(t) - C_A(\infty) \propto t^{-d/2} \tag{8}$$

Summing up the results on the long-time behavior of reversible reactions, we conjecture[29–31] that a power-law approach to equilibrium is valid for all reversible reactions, since for an set of successive and parallel reactions there always exists a pure diffusive mode—a linear combination of local concentrations not affected by chemical conversions (e.g., the sum of reagent and product local densities).

However, it should be mentioned that in the simplest systems considered above, with random homogeneous, mutually independent initial particle distributions without correlations and with small mean concentrations, fluctuations influence only the long-time kinetic tails. This means that a very small amount of particles C_f [$C_f = C(t_f)$, t_f a crossover time from the mean-field to the fluctuation-induced kinetic behavior] reacts in the fluctuation-induced regime. For SB reactions in Eq. (1a), $\ln[C_f/C(0)] \propto \ln \beta$ and for the trapping problem $\ln \ln[C_f/C(0)] \propto \ln \beta$, where β is the initial fraction of reactants, $\beta = R^d \max[C_A(0), C_B(0)]$. Here we have a straightforward analogy with second-order phase transitions in systems with small coupling, in which fluctuations are important in the narrow region near the critical point. The latter is the analog of the asymptotic time regime in DCP problems.

4. Correlation effects are essentially enhanced in systems prepared by a steady external source.[32–36] If the particles A and B are generated

randomly and independently of each other at a constant average rate, the irreversible reaction in Eq. (1a) (and also the reversible one[29,30]) leads to the appearance of strong correlations in the distribution of species. The long-wavelength asymptotic behavior of the fluctuation spectrum changes[32-35]—it is not flat. Interestingly, the approach of $C_A(t)$ to equilibrium and the formation of the fluctuation spectrum obey the fluctuation-induced law $C_A(\infty) - C_A(t) \propto t^{-1/2}$ in $d = 3$ (see ref. 34), in place of the exponential mean-field prediction. The existence of an essential peculiarity in the steady-state fluctuation spectrum leads to a change in the long-time kinetics of Eq. (1a) after the source is switched off,[32] $C_A(t) \propto t^{-1/4}$ for $d = 3$. As in the systems considered earlier, nontrivial relaxation laws and the long-wavelength peculiarities of spatial correlates are caused by a balance between fluctuations of $z(r, t)$ generated by the random source of particles and diffusive smoothing. It is important to note that in low-dimensional systems ($d \leqslant 2$), with arbitrary relations between the rate constants, the diffusive processes fail to equalize the spatial inhomogeneities. This leads to a spontaneous separation of homogeneous (on average) systems into macroscopic domains containing particles of only one type.[32-37] Aside from this, the average concentration increases with the generation time. These effects have also been observed numerically[35] and predicted theoretically[37,38] for reactions on fractals.

A similar picture is evident in low-dimensional systems with parallel reactions—recombination ($A + B \rightarrow 0$), slow annihilation ($A + A \rightarrow 0$, $B + B \rightarrow 0$), and multiplication ($A \rightarrow nA$, $B \rightarrow nB$, $n > 1$). In the course of reactions the system divides into domains of random linear sizes. After this, the larger domains begin to swallow their smaller neighbors. Both the growth of the average linear domain size and the evolution of mean concentrations are governed by slow logarithmic laws.[39]

It also appears that fluctuation effects are strongly enhanced by multiplication reactions. For instance, a trapping reaction in Eq. (1c) in 3D, followed by the multiplication of A, exhibits unusual behavior if the mean trapping rate $4\pi a D C_T$ is larger than the multiplication rate constant k_m. In such systems C_A drops exponentially, i.e., in accord with the mean-field theory law

$$C_A(t) = C_A(0) \exp[-(4\pi a D c_T - k_m)t]$$

After the concentration decreases to the small value $C_A(0) \times \exp(-\text{const}/\sqrt{\beta})$, it remains approximately constant for a long time, after which it grows as $\exp(k_m t)$ (see ref. 14). Hence the kinetic behavior is not monotonic.

Fluctuation effects are also very important in reactive systems with nondiffusive random motion and distant reactions, characterized by trans-

fer rates $\omega(r)$ dependent on the mutual distance between reactants. For multipolar transfer (MT), $w(r) \propto r^{-s}$, and for exchange-mediated transfer (ET), $\ln w(r) \propto -r$. Let us consider the averaged survival probability $P(t)$ of excitations migrating along a disordered system consisting of donors and accepters which permit quenching. It can be shown[40] that in a random field that determines the probability of quenching, the averaged survival probability can be only decreased by the random motion provided that the latter is uncorrelated with random field realizations. On the basis of this simple statement and rigorous lower bounds we have obtained[40] exact result for $P(t)$ at $t \to \infty$: $\ln P(t) \propto t^{-d/s}$ for MT, and $\ln P(t) \propto -\ln^d(t)$ for ET. This survival probability is supported by excitations which do not move from the initially excited donors. It has been also shown that for a wide class of transfer rates and concentrations the fluctuation-induced kinetics is valid even at early stages of the process.

In ref. 41 we have studied the kinetics of static recombination (or "direct reaction"; see refs. 42 and 43) in a system of randomly distributed immobile particles of two types $(A + B \to C)$. In this we have dealt only with the case of equal mean concentrations: $C_A(0) = C_B(0)$. We have also analyzed a system of particles of one type $(A + A \to C)$. Since the smoothing influence of diffusion is absent, in this case one expects that the role of fluctuations is even more essential than in earlier cases.

Let us consider a system in which A and B particles are initially located at random points $\{r_i^A\}$ and $\{r_j^B\}$. The probability dp for two particles A_i and B_j to react and be removed from the system in time interval dt is determined by a transfer rate $w(r_{ij})$ that depends on the mutual distance r_{ij}. The probability for an isolated pair (A, B) not to react until moment t is given by $\tau^{(0)}(t) = \exp[-w(r) \cdot t]$. The survival probability $\tau_i^A(t)$, i.e., the ensemble-averaged probability for a given particle A_i to survive until moment t, satisfies the set of equations

$$\frac{d\tau_i^A}{dt} = -\tau_i^A W_i = -\tau_i^A \sum_j \tau_j^B w(r_{ij}) \tag{9}$$

where W_i is the ensemble-averaged total transfer rate of particle A_i (the particle A_i can react with the particle B_j only in case both of them survived). Turning to the particle concentration $C_A(r, t) = C_A(r, 0) \cdot \tau^A(r, t)$, we obtain from Eq. (9)

$$\dot{C}_A(r, t) = -C_A(r, t) \int w(r - r') \, C_B(r, t) \, dr' \tag{10}$$

with a similar equation for $C_B(r, t)$.

The mean-field approximation, which corresponds to neglecting correlations between the particles of the same type, yields the asymptotic law

$$C(t) \propto \ln(t)^{-d} \tag{11}$$

The mean-field decay law in Eq. (11) describes correctly the behavior of a system with particles of a single type $(A + A \to C)$.[14] The fluctuation decay law for the (A, B) system with the ET rate

$$C(t) \propto \ln(t)^{d/2} \tag{12}$$

has been suggested and explained in ref. 14. It has been confirmed by the numerical solution of Eqs. (9) within the framework of the Kirkwood approximation for correlation functions, as well as having been tested by simulations.[44]

In ref. 41 it was proved that for reaction $A + B \to C$ and the exchange transfer rate the result of Eq. (12) represents the exact decay law, and for the multipolar transfer rate a new exact result has been obtained,[41]

$$C(t) \propto t^{-d/(2s-d)} \tag{13}$$

(s should be greater than d). It has also been proved that the mean-field decay law in Eq. (11) for the exchange transfer rate, and the mean-field decay law

$$C(t) \propto t^{-d/s} \tag{14}$$

for the multipolar transfer rate, describe the kinetics of $A + A \to 0$ reactions correctly.

Reaction kinetics are drastically changed in systems with a high volume fraction of reactants (above the percolation threshold), in polymer solutions, and in crystals with topological defects.[46] For these, the correlation effects are important even at earliest times. Let us consider the kinetics of Eq. (1c) for two percolationlike systems for which, in the absence of a reaction, the A particles are localized in finite volumes. The A particles, each of a charge q, diffuse in the presence of an external uniform electric field on a lattice, whose sites can be occupied by immobile, randomly distributed, entanglements S. It was shown[45,46] that if there exists a slow trapping of A by S, the reaction kinetics goes on as follows. In the absence of an electric field the long-time behavior is governed by the correlation-induced law given in Eq. (3). However, intermediate kinetics, which defines the annihilation of the bulk of A, is also not properly given by the mean-field result, but rather by

$$\ln C_A(t) \propto -t^{d/d+1} \tag{15}$$

In an electric field the decay in Eq. (15) also defines the intermediate kinetics, but the long-time decay follows an exponential dependence upon time

$$\ln C_A(t) \propto -Et \quad \text{and} \quad \ln C_A(t) \propto -E^2 t \tag{16}$$

for $E \gg E_{cr}$ and $E \ll E_{cr}$, where E_{cr} is a critical value of the external field. In the second case, particles S are neutral with respect to the reaction and the decrease of A occurs in an encounter with particles of a third species T, where trapping centers are randomly distributed on a lattice among the sites not occupied by particles of species S. In the limit $t \to \infty$, $C_A(t)$ tends to a constant value $C_A(\infty)$, which is equal to the fraction of localization cavities which contain no trapping centers. In the limit $t \to \infty$ we find[45,46]

$$
\begin{aligned}
\ln[C_A(t) - C_A(\infty)] &\propto -t^{1/2} \quad &(E = 0) \\
\ln[C_A(t) - C_A(\infty)] &\propto -\alpha \ln t \quad &(E > 0)
\end{aligned}
\tag{17}
$$

where α is a nonuniversal constant dependent on E and concentrations.

5. The course of reaction kinetics for disordered systems is often completely determined by the distribution of diffusion fluxes. In this last section we present some results concerning the steady-state diffusive flux for two models—the membrane containing randomly distributed repulsive impurities and the finite disordered 1D lattice with random nonsymmetric transition probabilities (the Sinai chain). We show that fluctuations are important in both cases and the steady-state flux is non-Fickian.

In ref. 47 we investigated the steady-state permeability $\chi(L)$ of a three-dimensional barrier membrane containing randomly distributed immobile repulsive impurities as a function of the membrane thickness L. The diffusive permeability of the membrane is defined as

$$\chi = \frac{L}{D \, \varDelta C_{ph}} \langle J \rangle \tag{18}$$

where D is the diffusion coefficient in the membrane without impurities, $\varDelta C_{ph}$ is the difference between substance concentrations in phases outside of the membrane on both sides of it, and $\langle J \rangle$ is the mean diffusive flux (averaged over different realizations of the distribution of impurities).

We have demonstrated that fluctuations in the distribution of the impurities enhance the value of $\chi(L)$ and cause anomalous behavior compared to Fick's law. For thin membranes $(L < L^*)$ the permeability χ falls off as $\exp(-\text{const} \cdot L)$; for intermediate thicknesses $(L^* < L < L^{**})$ the stretched-exponential dependence is valid, $\chi \propto \exp(-L^\gamma)$, $\gamma < 1$; and for

$L > L^{**}$ the permeability χ approaches a constant value, i.e., Fick's law is valid. The borderline thicknesses L^* and L^{**} are functions of the temperature ($L^* \to \infty$ and $L^{**}/L^* \to \infty$ as $T \to 0$), the impurity concentration, and the parameters of the repulsive potentials. Analogous dependences were predicted for the steady flux through the barrier membrane containing randomly distributed hard-core obstacles.[48]

For the one-dimensional Sinai chain the random walk is defined as follows: at each site j there is a probability p_j to hop to site $j + 1$ and probability $q_j = 1 - p_j$ to hop to site $j - 1$. The set $\{p_j\}$ consists of independent random variables, which satisfy

$$\langle \ln[p_j/q_j] \rangle = \int \rho(p_j) \, dp_j \ln[p_j/(1 - p_j)] = 0 \qquad (19)$$

where the angle brackets refer to the disorder average with the distribution $\rho(p)$. Sinai[49] has proved rigorously that the mean-square displacement of a walker in such a system is proportionally to $\ln^4 t$, t being the time. This remarkable slowing down of the diffusion process, known as Sinai diffusion, or random walk, has led to a large number of investigations.

It has been suggested that Sinai diffusion might be relevant to various physical phenomena, such as $1/f$ noise,[50] the slow dynamics of random field magnets,[51] the dynamics of dislocations in doped crystals,[51] and anomalously slow diffusion of particles constrained to move on a random linear structure in an external field.[52] For more details there are exhaustive reviews on this subject.[51,53]

In ref. 54 we have studied the disorder average steady flux $\langle J(N) \rangle$ in a one-dimensional Sinai chain as a function of chain length N, a property which actually depends on a new class of a typical realizations of $\{p_j\}$. We have shown that in a Sinai-type disordered chain $\langle J(N) \rangle \geqslant 1/\sqrt{N}$ (with possible logarithmic corrections) for sufficiently large values of N, i.e., the steady flux in such a disordered system is much greater than the usual Fickian flux $J(N) \propto 1/N$. The numerical data strongly support the dependence $\langle J(N) \rangle \propto 1/\sqrt{N}$. The exact solution of the continuum-space analog of this problem leads[55] to the same dependence on N.

6. CONCLUSION

One can qualitatively explain the results discussed in this report in a very simple manner.[56]

For the diffusion-controlled reaction $A + A \to 0$ fluctuation effects are not essential. The rate of this reaction increases with an increase in local concentration decreases. Therefore, fluctuation are suppressed[23] and the

mean-field theory leads to exact results.[16] If the random migration is sub-diffusive, a special type of fluctuation (odd–even number of particles) is important. Let us denote the characteristic displacement by $L(t)$. In a wide set of reactive systems $L(t) \propto t^{\nu}$. For the slow motion when $d\nu < 1$ (so-called compact exploration[57]) in the volume $V \propto L^{d}$ no A particle survives if the initial number of A's is even. When the initial number of particles is odd, then the average number of survivors is equal to unity, which leads to the kinetic law $C_A(t) \propto L^{-d} \propto t^{-d\nu}$.

In the course of the reaction $A + B \rightarrow 0$ the ratio of the characteristic fluctuations to the mean concentration increases.[23] The mean concentration in the volume considered is completely determined by the excess of A or B particles. Consequently, for random mutually independent A and B initial distributions, $C_{A,B}(t) \propto L^{-d/2}$, which leads to the decay of Eqs. (5), (6), and (13).

For reversible reactions with the same initial distributions the relaxation of the mean concentration has the dependence $\delta C \propto L^{-d/2}$. In general, the power in these decay laws is a functional of features of the large-wave fluctuation spectra.[29,30,40]

In diffusive motion, $\nu = 1/2$. In static recombination, the random walk in random environment,[4] polymer systems,[57] diffusion in a 1D hard-core lattice gas, and many other important systems, the value of ν is less than $1/2$. This leads to a wide variety of nontraditional kinetic laws.

The kinetic behavior of recombination and annihilation reactions is determined by typical small fluctuations. In contrast, the survival probability in the trapping problem and averaged flux in systems with strong disorder are based on untypically large fluctuations. The probability of a given fluctuation decreases with an increase in its magnitude, but its contribution to the survival probability increases. As a rule, the most significant fluctuation value is determined by the steepest descent method. It essentially depends on correlations in the reactive system.

To summarize, we have presented some results corresponding to many-particle kinetics of reactions in condensed media. We have shown that the kinetic behavior can be drastically changed due to fluctuation effects, spatial correlations between particles, and anomalous transport properties. Moreover, we have shown that the correlation-induced kinetics in some systems can govern the density evolution over the entire time domain.

ACKNOWLEDGMENT

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